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The primary goal of the proposed research is to study and develop alloying concepts for understanding intermetallic alloys as derived from a first principles quantum mechanical approach. Thus, a major part of our effort will be to study and determine ductilizing effects of alloying elements in several intermetallic compounds and to work closely with experimental efforts to evaluate the applicability of the theoretical approach to alloy design. Specifically, highly precise all-electron quantum mechanical electronic structure methods will be applied to the study of a number of materials problems in order to obtain from first principles information of relevance to alloy stability and the design of structural materials. Using our recently developed state-of-the-art all-electron self-consistent total energy methods which give precise solutions of the local density equations, fundamental information will be sought about the structural and electronic properties of these alloys in order to predict stable and metastable phases and how alloying affects bonding, crystal ordering and crystal symmetry. The first principles approach used here will address questions of a metallurgical nature, such as phase stability, crystal structure, equilibrium lattice constants, and mechanical properties including the effect of atomic relaxation. The proposed research seeks to explore a new capability for modeling materials and their properties on the computer which have not yet been made in practice.

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PUBLICATIONS:

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"Electronic Structure and Mechanical Properties of Intermetallics: APB Energies in Ni-Al-based Systems", by T. Hong and A.J. Freeman, MRS Mtg., Boston, MA, Nov. 1988.

"Structural Stability and Elastic Properties of Al_3Li Intermetallic Compounds", by X. Guo, A.J. Freeman, and R. Podloucky, APS Mtg., St. Louis, MO, March 20-24, 1989.

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"Bandfilling and Structural Stability of Transition Metal Trialumindies: MAl_3", by J.H. Xu and A.J. Freeman, submitted to APS Mtg., Anaheim, CA, March 12-16, 1990.

"Twin Boundary and Intrinsic Stacking Fault Energies of Al and Pd", by J.H. Xu and A.J. Freeman, submitted to APS Mtg., Anaheim, CA, March 12-16, 1990.

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ABSTRACT OF OBJECTIVES AND ACCOMPLISHMENTS:

The primary goal of the proposed research is to study and develop alloying concepts for understanding intermetallic alloys as derived from a first principles quantum mechanical approach. Thus, a major part of our effort will be to study and determine ductilizing effects of alloying elements in several intermetallic compounds and to work closely with experimental efforts to evaluate the applicability of the theoretical approach to alloy design. Specifically, highly precise all-electron quantum mechanical electronic structure methods will be applied to the study of a number of materials problems in order to obtain from first principles information of relevance to alloy stability and the design of structural materials. Using our recently developed state-of-the-art all-electron self-consistent total energy methods which give precise solutions of the local density equations, fundamental information will be sought about the structural and electronic properties of these alloys in order to predict stable and metastable phases and how alloying affects bonding, crystal ordering and crystal symmetry. The first principles approach used here will address questions of a metallurgical nature, such as phase stability, crystal structure, equilibrium lattice constants, and mechanical properties including the effect of atomic relaxation. The proposed research seeks to explore a new capability for modeling materials and their properties on the computer which have not yet been made in practice.

I. INTRODUCTION AND BACKGROUND PERSPECTIVE

The high temperature intermetallics have become a major focus of efforts - both experimental and theoretical - driven in large part by their potential for important aerospace applications. In the theoretical area of this research, our work has been at the forefront and has already stimulated similar efforts at other laboratories. Indeed, the work initiated and carried out under AFOSR support reflects the present state of the field and the reaching of a threshold: advanced theoretical - computational techniques combined with the power of supercomputers provide an understanding of matter at the atomic-scale with an unprecedented level of detail and accuracy. It is this capability that has given birth to a new branch of scientific endeavor: Computational Materials Science. Not only can we simulate experiment, but using the computational approach, we are now beginning to design new materials and to predict their properties without actually synthesizing them. In contrast to an analytic-theoretical approach, which isolates and idealizes real systems to unravel fundamental relations and laws, the computational approach is synthetic: its goal is to simulate more and more details of the systems studied including as much of the environment as possible.

Designing new materials with specific mechanical, thermal, chemical, electronic, magnetic and optical properties hinges on one basic assumption: the properties of the macroscopic ensemble are related to and can be derived from the properties of individual molecules and atomic building blocks (such as crystallographic unit cells in solids). But which atomic-scale physical quantities and observables need to be looked at? This is the important question today and the key to successful materials by design in the future.

Today much of the theoretical efforts are focused on the atomic scale since the theoretical concepts and methodologies being developed are bringing us closer to the goal of a unified approach. The theoretical approach, density functional theory, that underlies all the calculations discussed here is appealing because it promises to provide such a unified approach, which is particularly suited for bridging the gap between our understanding of atomic scale properties and the macroscopic properties essential for producing useful materials applications.

This proposal seeks continued support for what has become a prolific and innovative research program. (See Appendix A for a list of publications reporting our accomplishments.) This report focusses on our recent efforts to understand, from first principles, a number of properties of aluminum intermetallic alloys important for potential aerospace applications. These center around the key issues of bonding, cohesion and phase stability of the intermetallics and the role of ternary additions on their structural, electronic and mechanical properties.

1. ELECTRONIC STRUCTURE AND PHASE STABILITY IN TRANSITION-METAL TRIALUMINIDES

We have studied the electronic structure and the structural stability of $TMAl_3$ (TM=Sc,Ti,V,Y,Zr and Nb) in the $L1_2$, the DO_{22} and the naturally stable form (i.e., DO_{19} and DO_{23} phases for YAl_3 and $ZrAl_3$, respectively) using the total energy local-density approach[1].

We have calculated the total energy as a function of the Wigner-Seitz(WS) sphere radius for $ZrAl_3$ in the $L1_2$, DO_{22} and DO_{23} structures. We find that the locations of the equilibrium r°_{ws} are very close to each other. For all these trialuminides, the calculated energetically favored structures are completely in agreement with the experimentally observed stable structures.

It is interesting to note that the differences in E_{tot} (~14-19 mRy/f.u.) for the group IV TM trialuminides(TiAl$_3$ and ZrAl$_3$) between the stable and hypothetical phases are much smaller than those(~72 mRy/f.u.) for both group III TM{ScAl$_3$ and YAl$_3$) and group V TM(VAl$_3$ and NbAl$_3$) trialuminides. This indicates a crossover situation between the stable and the hypothetical phases. Therefore, ternary additions in the group IV TM trialuminides may alleviate the phase transition amoung the different phases. This proves indeed to be the case; the additions of Cu, Ni, Fe, Mn, Cr, Zn and Ag to TiAl$_3$ yield L1$_2$ compounds (such as, Al$_5$CuTi$_2$) [2], and partial substitution of V for Zr in ZrAl$_3$ increases the thermal stability of the metstable L1$_2$ phase in dulite alloys [3]. By contrast, it has not been possible to convert DO$_{22}$ into L1$_2$ by means of ternary additions[4] to NbAl$_3$, which can be understood from the large E_{tot} difference (~ 72 mRy/f.u.) between the stable (DO$_{22}$) and hypothetical (L1$_2$) phase. In particular, note that the calculated results provide a very clear trend of increasing(decreasing) structural stability of the DO$_{22}$(L1$_2$) phase on going from YAl$_3$ to ZrAl$_3$ to NbAl$_3$. Likewise, we have seen the same variation of the structural stability from ScAl$_3$ to TiAl$_3$ to VAl$_3$. In addition, note that the stable phase is often accompanied with a lower N(E$_F$) [5].

In order to gain insight at the microscopic level into the phase stability of the TM trialuminides and its variation with the TM constituents, we also investigated the electronic structures of the three trialuminides YAl$_3$, ZrAl$_3$ in the L1$_2$ and DO$_{22}$ structure. For example, an analysis of the Nb-4d and Al-3p partial density of states (DOS) for DO$_{22}$ NbAl$_3$, demonstrates that the hybridization between the Nb-d and Al-p states is so substantial that the Nb-d and Al-p states almost overlap each other completely in the whole energy region from the bottom of the band to high above E$_F$. Further, a broad and prominent peak formed at about -1.1 eV for YAl$_3$, -1.5 eV for ZrAl$_3$, and -2.1 eV for NbAl$_3$ arises mainly from the TM-d - Al-p bonding states and a deep valley (or pseudogap[6]) between the bonding states and the nonbonding states is located at about 1.6 eV for YAl$_3$, 1.1 eV for ZrAl$_3$. We find that the overall features of the total DOS for these three trialuminides resemble each other. Similarly, we have nearly the same DOS curve for ScAl$_3$ and TiAl$_3$ and VAl$_3$. In other words, the rigid band approximation holds well for these trialuminides when we move from group III to IV to V TM. As a result, the valence electrons gradually fill the bonding states, the Fermi level for YAl$_3$ shifts up to ZrAl$_3$, and finally moves close to the pseudogap in the DOS for NbAl$_3$(VAl$_3$). Thus, one is led to a phase transition from the Ll$_2$ phase for YAl$_3$(ScAl$_3$) to DO$_{22}$ for NbAl$_3$(VAl$_3$) (in a broad sense, Jones' explanation for the Hume-Rothery rule [7]).

2.   ELECTRONIC STRUCTURE AND MECHANICAL PROPERTIES

A.   Anti-Phase Boundary Energies in Off-stoichiometric FeAl and NiAl

The B2 aluminides, FeAl and NiAl, have been studies as high temperature structural materials for more than two decades due to a number of intriguing properties [8,9,10,11,12,13,14]. From a number of experimental studies, the major deformation mode in stoichmetric B2 NiAl was found to be <001> slip [15]. With only three independent slip systems, the <001> slip could not meet the von Mises [16] criterion for a uniform, homegeneous plastic deformation in polycrystalline materials. On the other hand, stoichiometric FeAl deforms mainly by <111> slip [12, 17], which could support ductile deformation as the von Mises criterion is now met. Further studies showed that the brittleness in FeAl could be attributed to interstitial impurities and microcracks left by machining [18], instead of the intrinsic factors as in NiAl. Hence, it is speculated that the activation of <111> slip in NiAl might change its brittle nature.

Since the APB energy is an important factor in controlling slip behavior [19] and little work has been done on the influence of ternary elements on APB energies [10], application of the state of art first principles calculations to these problems became a natural step. In an erlier study, we presented results of calculations of APB energies for stoichiometric NiAl with selected ternary additions[20]. Extremely high APB energies for stoichiometric NiAl and remarkably reduced values for NiAl with a few ternary substitutions were obtained. For comparison and for it's own interest, the calculation of APB energies for the similar B2 compound FeAl is of great importance.

Ray, et al. [21] estimated the APB energies for a few compositions of B2 FeAl in the Fe rich side of the phase diagram and obtained decreasing APB energies with increased Fe content. Later studies by Mendiratta, et al. [22,23,24] also indicated the tendency to ease <111> slip with decreasing Al content. More recently, Munroe and Baker[25] observed <111> slip along with <001> slip in an extruded sample of $Ni_{0.58}$ $Al_{0.42}$. Thus, a systematic investigation of the effect of stoichiometry on the APB energies for B2 FeAl and NiAl could help understand the mechanism behind the slip behavior. In this section, we present the results from calculations for stoichiometric B2 FeAl as well as $Fe_{0.6}Al_{0.4}$ and $Ni_{0.6}Al_{0.4}$.

To explore the effect of the location of excess Fe (Ni) atoms on the APB energies, we considered three different configurations which all correspond to the composition with 40 atomic percent, Al, but with different location of excess Fe(Ni) atoms relative to the APB's. $M_{12}Al_8$(I) and $M_{12}Al_8$(II) correspond to the cases in which the excess M atoms are located in the second and the first (110) layer away from the possible APB interface, respectively. $M_{12}Al_8$(III) is the case in which the excess M atoms are located right in the possible APB interface.

By comparing the total energies of the B2 with and without APB, the APB energy for stoichiometric FeAl was found to be about 500 mJ/m^2. This is to be compared with (i) the crude estimate of Mendiratte, et al.[24] (175 mJ/m^2) using Potter's scheme [26], (ii) the unpublished result by Inden [27] (160 mJ/m^2 and (iii) the extrapolated value (233 mJ/m^2) from the work of Crawford, et al.[28]. The discrepancy is probably due to the crudeness of the earlier estimates as well as the neglect of atomic relaxation at the APB interface in our calculations.

The calculated APB energies for FeAl from the supercells with 12 atoms (6 layers) and 20 atoms (10 layers) only differ by about 4%, compared to about 12% for NiAl [20]. This indicates a faster convergence rate for the APB energy with respect to the number of layers between two consecutive APB's in FeAl than in NiAl, which probably implies weaker interactions in FeAl than in NiAl.

By comparing the total energies of $M_{12}Al_8$ (I,II,III) supercells with and without APB's, three APB energies for $Fe_{0.6}Al_{0.6}$ ($Ni_{0.6}Al_{0.4}$) were obtained corresponding to three different APB locations relative to the excess Fe (Ni) atoms. It is very interesting to see the large difference in these three APB energies for both $Fe_{0.6}Al_{0.4}$ and $Ni_{0.6}Al_{0.4}$. The APB energies with the first and second choices of site occupancy for excess Fe (Ni) atoms are not changed much from the corresponding stoichiometric cases, showing an insensitivity of the APB energy in both B2 FeAl and B2 NiAl to changes in layers other than the APB interface. By contrast, in this last case (III), the APB energy is reduced by more than one order of magnitude from the stoichiometric value; this gives rise to an even greater effect than the reduction of the APB energy in B2 NiAl with ternary additions [20].

These results for different site occupancy of the excess F (Ni) atoms once again emphasize the dominance of the local environment in determining the electronic structure and associated mechanical properties. It is clearly seen that the APB energy depends predominantly on the net changes of the first and second nearest neighbors in the supercell. With greatly reduced APB energies, the activation of <111> slip is

obviously eased in the region where excess Fe (Ni) atoms are located. For the region with composition near stoichiometry, however, the ease of activating <111> slip remains about the same as that for stoichiometric FeAl (NiAl). Thus, the effect of the excess Fe (Ni) atoms is limited only to the local excess region, as far as the creation of APB's is concerned. Based on these results, we propose a simple model to explain the observed linear dependence of the APB energy in B2 FeAl on deviation of the composition from stoichiometry [21,28]. As the deviation from stoichiometry increases, the portion of the region with excess Fe (Ni atoms grows linerly, thereby rendering a linearly decreased average APB energy in B2 FeAl which was obtained experimentally also as the statistical average over a large number of measurements [28].

We expect that the same model should also be applicable to B2 NiAl, although there are no experimental results to date for the compositional dependence of the APB energy due to the unavailability of APB's (which is consistent with the extremely high APB energies from our calculations [20]). However, the experimental studies of Munroe and Baker [25] (mentioned above) could surely support such a model. If this model were suitable over the whole B2 composition range, the APB energy would be at the lowest value for the alloy with composition right at the B2 phase boundary on the decreased Al side.

B. APB Energies and Bonding Characteristics in RuAl and NiAl

RuAl is another aluminide possessing the highly symmetric B2 structure. It has an even higher melting temperature (2060°C) than NiAl (1638°C). In RuAl, a ductility in compression was reported [29] contrary to the severe brittleness NiAl suffers at ambient temperature. The slip vectors of RuAl were found to be in the <100>, <110> and <111> directions, which provides enough slip systems. In NiAl, in addition to the high anti-phase boundary(APB) energy associated with the <111> direction, it is speculated that the absence of the slip vector in this direction could be caused by a slight excess charge in <111> [14]. A covalent type of bonding was proposed [30,31].

Cohesive properties and APB energies of RuAl and NiAl were studied. The equilibrium lattice constants calculated by the LMTO method for both compounds were found to be in very good agreement with the experimental values. For NiAl the difference is less than 0.5% (2.89 Å vs 2.886 Å[32]) and the calculated result for RuAl is exactly the same as experimental result [33] (3.03 Å). The lattice constant of NiAl and RuAl calculated by the FLAPW method are about 1.0% ~ 1.7% smaller than the experimental value. The results show both NiAl and RuAl to have a large bulk modulus. For NiAl it is about 10% larger than experiment; for RuAl, no experimental result is available at present. NiAl has been experimentally found to have a large formation energy (29.8 kcal/mol), and larger than its closest neighbor aluminides such as CoAl (25.8 kcal/mol) and and FeAl (12.0 kcal/mol). Our calculation gave results which are within 15% of experiment. The formation energy for RuAl was calculated to be fairly large (35.8 kcal/mol by LMTO and 36.2 kcal/mol by FLAPW; both values are much larger than that for NiAl). No experimental data is available for direct comparison.

Only a six-layered (12 atom) supercell was used to obtain and compare the APB energy of RuAl and NiAl. The results show that NiAl has a very large APB energy (1000 mJ/m^2) in the <111> {110} direction (calculated from the six-layered supercell). On the other hand, the APB energy of RuAl is only 690 mJ/m^2, or 70% that of NiAl. The estimated reduction of the APB energy is 80% after expanding the size of the supercells. This leads to an estimated APB energy near 550 mJ/m^2 for RuAl - a result comparable to that for FeAl (about 490 mJ/m^2) calculated recently[20] whose dislocation in that direction is proven to exist[34] and TiAl (510 mJ/m^2)[35].

The Ru d band in RuAl is much broader than is the Ni d band in NiAl, but the DOS characteristics are similar to those of NiAl. The DOS profile and the location of E$_F$ agrees

with earlier work[36]. The Ru-d and Al-p hybridization is very strong below the Fermi energy which lies in the bonding region. In the partial DOS, the main peak below E_F is dominated by Ru-d electrons; the Ru-s, p electrons make only a small contribution. On the other hand, between -11.0 eV and -5.0 eV there is a sharp Al-s peak. Between -5.0 eV and 1.5 eV, Al-p and Al-d have comparable contribution.

C. Effect of Ternary Addition: $Al_3Ti + Cu$

Among intermetallic compounds, titanium-aluminides are considered as some of the most promising candidates for aerospace applications and to have superior properties to the Ni-based superalloys [37]. Since Al_3Ti crystallizes in its stable DO_{22} structure, i.e., and $L1_2$ derivative with tetragonal symmetry, it is natural to try to stabilize the closely related cubic $L1_2$ structure which could offer more slip systems and, consequently, might make the material more ductile.

So far, a number of ternary elements (such as Cu, Ni,Fe and Zn) have been found to promote the $L1_2$ structure in Al_3Ti. As Cu is the element with wide composition range [38] in Al_3Ti and was reported to form the compound Al_5CuTi_2[39] (which other elements don't), we chose $Al_3Ti + Cu$ for our first principles calculations. Further, since Al_5CuTi_2 was observed experimentally, we only considered a composition of 12.5% Cu to replace either Al or Ti in Al_3Ti. By doing the substitution for both DO_{22} and $L1_2$ Al_3Ti, we obtained four different unit cells (configurations): $L1_2$- and DO_{22}-like Al_5CuTi_2- and DO_{22}-like Al_6CuTi. To examine the effect of varying c/a ratios for these tetragonal cells, we performed calculations at two characteristic c/a values, one at c/a=2 which is related to the undistored $L1_2$ structure, the other at c/a = 2.234 which is the observed value for DO_{22} Al_3Ti [32].

Our results indicated that Cu as a ternary addition strongly favors the Al site (by about 0.2 eV/atom) over the Ti site (for c/a = 2 case). In contrast to the earlier calculations for binary Al_3Ti [40,41], the tetragonal distortion does not reverse the relative stability of the $L1_2$- and DO_{22}-like structures, although the energy difference changes with different c/a values. Except for $L1_2$-like Al_5CuTi_2, where the formation energy at c/a = 2.0 is about 0.07 eV/atom lower than that at c/a = 2.234, the other structures favor a tetragonal distortion. The $L1_2$-like structure is obviously stabilized when Cu is added to Al_3Ti with a more than 0.1 eV/atom larger formation energy than that of the DO_{22}-like structure. Our calculated site preference and relative stability of the $L1_2$- and DO_{22}-like structures all agree with experiment. Futhermore, our results indicate that the choice of site occupancy is more important than the structural difference.

The density of states obtained for Al_5CuTi_2 and Al_6CuTi revealed two different effects. As we suggested in an earlier paper [42], the rigid band model works very well for Al_5CuTi_2. The profile of the total DOS for Al_5CuTi_2 is very much like that in pure Al_3Ti except for a prominent peak associated with Cu d electrons which are basically not hybridized with other components. Since Cu has fewer electrons participating in bonding than either Ti or Al, the Fermi energy is shifted towards lower energy in both the $L1_2$ and D_{22}-like structures compared to their counterparts in pure Al_3Ti. The number of Cu electrons provided is just right to put E_F in a deep valley separating bonding from non-bonding and antibonding regions for the $L1_2$ structure. On the other hand, fewer electrons in Al_5CuTi_2 put E_F of the DO_{22}-like structure near a major peak due to the hybridization of Al-p and Ti-d electrons. The non-bonding and antibonding contribution is totally unoccupied in the $L1_2$-like structure, while the bonding contribution is weakened in the DO_{22}-like structure, resulting in the stabilized $L1_2$-like structure over the DO_{22}-like structure.

By comparing the valence charge density for different configurations, it was found that the substitution of Cu for Al causes similar bonding character in the new material as the tetragonal distortion did in pure Al_3Ti. For pure Al_3Ti, the charge density in the region between the first nearest neighbor Ti-Al and the second nearest neighbor Ti-Ti or Al-Al on the (001) plane with equal numbers of Ti and Al atoms is generally larger in the DO_{22} structure than in the $L1_2$ structure. Meanwhile, the charge density in the DO_{22} structure along the bond directions connecting the atoms on different (001) planes (denoted as non-planar bonds) is still comparable with that in the $L1_2$ structure, resulting in a stronger hybridization for DO_{22} Al_3Ti when one counts all the bonds together. In $L1_2$-like Al_5CuTi_2, the charge density (with c/a = 2.0) on the (001) plane between the neighbors is larger than that in pure Al_3Ti (with c/a = 2.0), thus surprisingly showing a similar effect to that found as one introduces tetragonal elongation into pure Al_3Ti. On the other hand, the non-planar bonds in the $L1_2$-like Al_5CuTi_2 are still comparable to those in the $L1_2$ Al_3Ti, or at most, are slightly weakened from the latter. This tendency towards enhancing planar bonds in Al_5CuTi_2 can be used to explain why the formation energy for the $L1_2$-like structure at c/a = 2.0 is greater in absolute value than that at c/a = 2.234. As the point of tetragonal distortion is to maximize total bonding by adjusting the relative bonding strength of planar and vertical bonds and simultaneously keeping the volume constant, enhancing the planar bonds (forcing contraction in planar dimension) would weaken the vertical bonds (with accompanying elongation), and vice versa. Pure DO_{22} Al_3Ti prefers a tetragonal distortion (with observed c/a = 2.234) to maximize the bonding by enhancing the planar bonds.

In $L1_2$-like Al_5CuTi_2 (c/a = 2.0), although the tetragonal distortion does not occur geometrically, a "pseudo"-distortion is induced in the charge distribution by the ternary Cu atom to achieve a similar maximization of the bonding. When the similar (hypothetical) geometrical distortion as in pure Al_3Ti is added, it becomes excessive, as it would change the relative strength of planar and vertical bonds more than necessary to reach the delicate balance (for maximizing the total bonding). For Al_5CuTi_2 in the DO_{22}-like structure, the charge density features are similar to the $L1_2$-like case, but less strikingly so. Therefore, a tetragonal distortion is still effective in maximizing the bonding.

D. Interfacial Energy: Twin and Stacking Fault Energies of Al and Pd

Twin (TW) and stacking fault (SF) energies (in the broad sense, interfacial energies), are of significance for determining many physics properties. Thus, for example, it is well known that the mechanical properties are closely related to the existence of faults in alloys [43]. Despite the fact that a large amount of experimental data about fault energies have been accumulated for pure metals [44], there are still large uncertainties in the measured values of such interfacial energies. For example, the values of the TW energy for pure Cu range from 20 to 160 erg/cm^2. Direct measurements of these interfacial energies require very delicate techniques(such as the weak-beam electron microscopy method) to measure an extended singe dislocation. Moreover, the measured value of such an interfacial energy depends sensitively upon many factors, such as the existence of internal stresses, impurities in the matrix (which are experimentally extremely difficult to control) and temperature, etc.[43]

As an example of our emerging capabilities for dealing with such complex issues, we studied the (111) twin boundary and stacking fault energies of Al and Pd [44] using the all-electron total energy self-consistent linear muffin-tin orbital (LMTO) method within the framework of density functional theory. Fault energies were determined by

comparing the total energies obtained for the same size supercells for each of two cases (with and without fault).

The calculated TW and SF energies for Al and Pd are listed in Table I. The calculated intristic fault energy (280 ± 40 erg/cm^2) for Al has approximately the same value as the extrinsic one (~260 erg/cm^2). It is expected from the geometrical arrangement, i.e., in intrinsic SF (extrinsic SF) a layer is removed (inserted) into the otherwise normal sequence along the [111] direction. We do not understand the experimental trend, i.e., the extrinsic fault energy for Al is approximately 25% larger than the intrinsic one (cf.,Table I). Note that the calculated TW energy (130 ± 15 erg/cm^2) is in fairly good agreement with Ref. [46] (118 erg/cm^2) obtained from the LKKR method; however, it is in general 30% larger than that observed value (extrapolated to 0°K) for Al [44]. A plausible explanation for this discrepancy between the calculated and experimental values might be attributed to (i) neglect of the relaxation around the fault region, (ii) effect of the (finite) size of the supercell (or of neglecting the interactions between the two faults), and /or (iii) use of the local density approximation. (We should note that a similar overestimated value for the calculated antiphase boundary energy for NiAl [48]). Nevertheless, our calculated results appear to provide support for the experimental finding that $2E_{TW} \sim E_{SF}$ for both Al and Pd.

TABLE I.   Twin and stacking fault energies for Al and Pd (in erg/cm^2).

| | | Al | | | Pd | |
		TW	ISF	ESF	TW	ISF
	Ref. [45]	130±15	280±40	~260	97±5	-
Calc.	Ref. [46]	118	-	-	-	-
	Ref. [47]	61	160	133	-	-
	Ref. [43]	75	166	-	-	180
Expt.	Ref. [48]	75-100	135	180	-	-
0° K (extrapolated)	Ref. [44]	100-125	160	200	-	-

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